

# Resonant photon tunneling enhancement of the radiative heat transfer

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## Abstract

We study the dependence of the heat transfer between two semi-infinite solids on the dielectric properties of the bodies. We show that the heat transfer at short separation between the solids may increase by many order of magnitude when the surfaces are covered by adsorbates, or can support low-frequency surface plasmons. In this case the heat transfer is determined by resonant photon tunneling between adsorbate vibrational modes, or surface plasmon modes. We study the dependence of the heat flux between two metal surfaces on the electron concentration using the non-local optic dielectric approach, and compare with the results obtained within local optic approximation.

## 1 Introduction

It is well known that for bodies separated by  $d \gg d_T = c\hbar/k_B T$ , the radiative heat transfer between them is described by the Stefan-Boltzmann law:

$$S = \frac{\pi^2 k_B^4}{60 \hbar^3 c^2} (T_1^4 - T_2^4), \quad (1)$$

where  $T_1$  and  $T_2$  are the temperatures of solid 1 and 2, respectively. In this limiting case the heat transfer is connected with traveling electromagnetic waves radiated by the bodies, and does not depend on the separation  $d$ . For  $d < d_T$  the heat transfer increases by many order of magnitude due to the evanescent electromagnetic waves that decay exponentially into the vacuum; this is often referred to as photon tunneling. At present there is an increasing number of investigations of heat transfer due to evanescent waves in connection with the scanning tunneling microscopy, and the scanning thermal microscopy (STM) under ultrahigh vacuum conditions [1, 2, 3, 4, 5, 6, 7]. It is now possible to measure extremely small amounts of heat transfer into small volumes [8]. STM can be used for local heating of the surface, resulting in local desorption or decomposition of molecular species, and this offers further possibilities for the STM to control local chemistry on a surface.

The efficiency of the radiative heat transfer depends strongly on the dielectric properties of the media. In [3, 5, 6] it was shown the heat flux can be greatly enhanced if the conductivities of the material is chosen to maximize the heat flow due to photon tunneling. At room temperature the heat flow is maximal at conductivities corresponding to semi-metals. In fact, only a thin film ( $\sim 10\text{\AA}$ ) of a high-resistivity material is needed to maximize the heat flux. Another enhancement mechanism of the radiative heat transfer can be connected with resonant photon tunneling between states localized on the different surface. Recently it was discovered that resonant photon tunneling between surface plasmon modes give rise to extraordinary enhancement of the optical transmission through sub-wavelength hole arrays [9]. The same surface modes enhancement can be expected for the radiative heat transfer (and the van der Waals friction [10]) if the frequency of these modes is sufficiently low to be excited by thermal radiation. At room temperature only the modes with frequencies below  $\sim 10^{13}\text{s}^{-1}$  can be excited. For normal metals surface plasmons have much too high frequencies; at thermal frequencies the dielectric function of normal metals becomes nearly purely imaginary, which excludes surface plasmon enhancement of the heat transfer for good conductors. However surface plasmons for semiconductors are characterized by much smaller frequencies and damping constants, and they can give an important contribution to the heat transfer. Recently, enhancement of the heat transfer due to resonant photon tunneling between surface plasmon modes localized on the surfaces of the semiconductors was predicted in [7]. The authors studied the radiative heat transfer between a small particle, considered as a point-like dipole, and a flat surface. However, this treatment can

be applied for scanning probe microscopy only in the case  $R \gg d$ , where  $R$  and  $d$  are the radius of the particle and the separation between the particle and the surface, respectively. For the opposite limit  $R \ll d$ , which is more appropriate for the scanning probe microscopy, the heat transfer between the tip and the surface can, in the first approximation, be modeled by the heat transfer between two semi-infinite solids. In this case the multiple scattering of the electromagnetic waves by the surfaces of the bodies, which was not taking into account in [7], becomes important in the photon tunneling. In particular, at sufficiently small separation  $d$ , the photons go back and forth several times in the vacuum gap, building up coherent constructive interference in the forward direction much as would occur in resonant electron tunneling. In this case the surface plasmons on the isolated surface combine to form a "surface plasmon molecule", in much the same way as electronic states of isolated atoms combine to form molecular levels. This will result in a very weak distance dependence of the heat flux, because the transmission probability for photon does not depend on  $d$  in this case (see below). For large  $d$  the sequential tunneling is more likely to occur, where the photon excited in a surface plasmon mode, tunnels to the surface plasmon at the other surface, and then couples to the other excitations in the media and exit. Other surface modes which can be excited by thermal radiation are adsorbate vibrational modes. Especially for parallel vibrations these modes may have very low frequencies.

All information about the long-range electromagnetic interaction between two non-contacting bodies is contained in the reflection factors of the electromagnetic field. At present time very little is known about the reflection factors for large wave vectors and for extremely small frequencies. In our previous calculations of the radiative heat transfer and Van der Waals friction [11, 12, 5, 6] we mostly considered good conductors. In this case it was shown that the important contribution comes from the non-local optic effects in the surface region. However it was shown that the radiative heat transfer and Van der Waals friction becomes much larger for high resistivity material, for which the volume contribution from non-local effects is also important. Non-local optic refer to the fact that the current at point  $\mathbf{r}$  depends on the electric field not only at point  $\mathbf{r}$ , as it is assumed in the local optic approximation, but also at points  $\mathbf{r}' \neq \mathbf{r}$  in a finite region around the point  $\mathbf{r}$ . In the case when both points are located outside the surface region the dielectric response function can be expressed through the dielectric function appropriate for the semi-infinite electron gas. However, if one of the point  $\mathbf{r}$  or  $\mathbf{r}'$  is

located in the surface region, the dielectric response function will be different from its volume value, and this gives the surface contribution from nonlocality. In order to verify the accuracy of the local optic approximation we study the dependence of the radiative heat transfer on the dielectric properties of the materials within the non-local dielectric approach, which was proposed some years ago for the investigation of the anomalous skin effects [13].

## 2 Theory

The problem of the radiative heat transfer between two flat surfaces was considered some years ago by Polder and Van Hove [1], Levin and Rytov [2] and more recently by Pendry [3], and Volokitin and Persson [5, 6]. Polder and Van Hove were the first who obtained the correct formula for the heat transfer between two flat surface. In their investigation they used Rytov's theory [14, 15] of the fluctuating electromagnetic field. However, they presented their result only for identical media, and within the local optic approximation. In the subsequent treatment, they made numerical calculations not of the heat flux itself, but its derivative with respect to temperature, i.e., their result pertain only to the small temperature differences. Unfortunately, their paper contains no analytical formulas in closed form . Levin and Rytov [2] used the generalized Kirchhoff's law [15] to obtain an expression for the radiative heat transfer between two semi-infinite media in the impedance approximation. The case of the good conductors was investigated in the details both in normal and the anomalous skin effect region. Pendry [3] proposed a more compact derivation of the formula for the heat flux between two semi-infinite bodies due to evanescent waves, and calculated the heat transfer between a point- dipole and a surface. Volokitin and Persson [5] considered the problem of the heat transfer between two flat surfaces, as a particular case of the general approach for calculation of the heat transfer. They investigated numerically the dependence of the heat flux on the dielectric properties of the bodies, and found that for good conductors, even for very small distances the heat flux is dominated by retardation effects. They also showed that the heat flux heat is maximal at conductivities, corresponding to the semi-metal.

According to [1, 2, 3, 5] the heat transfer between two semi-infinite bodies , separated by a vacuum gap with the width  $d$ , is given by the formula

$$S = \int_0^\infty d\omega (\Pi_1 - \Pi_2) M \quad (2)$$

where

$$M = \frac{1}{4\pi^2} \int_0^{\omega/c} dq q \frac{(1 - |R_{1p}(\omega)|^2)(1 - |R_{2p}(\omega)|^2)}{|1 - e^{2ipd} R_{1p}(\omega) R_{2p}(\omega)|^2} + \frac{1}{\pi^2} \int_{\omega/c}^{\infty} dq q e^{-2kd} \times \frac{\text{Im} R_{1p}(\omega) \text{Im} R_{2p}(\omega)}{|1 - e^{-2|p|d} R_{1p}(\omega) R_{2p}(\omega)|^2} + [p \rightarrow s] \quad (3)$$

where the symbol  $[p \rightarrow s]$  stands for the terms which can be obtained from the first two terms by replacing the reflection factor  $R_p$  for the  $p$ -polarized electromagnetic waves with the reflection factor  $R_s$  for  $s$ -polarized electromagnetic waves, and where  $p = ((\omega/c)^2 - q^2)^{1/2}$ ,  $k = |p|$ . The Planck function of solid 1

$$\Pi_1(\omega) = \hbar\omega \left( e^{\hbar\omega/k_B T_1} - 1 \right)^{-1}, \quad (4)$$

and similar for  $\Pi_2$ . The contributions to the heat transfer from the propagating ( $q < \omega/c$ ) and evanescent ( $q > \omega/c$ ) electromagnetic waves are determined by the first and the second terms in Eq. (3), respectively.

Let us firstly consider some general consequences of Eq. (3). In the case of heat transfer through free photons ( $q \leq \omega/c$ ), the transfer is maximal when both bodies are perfectly black and have zero reflection coefficient,  $R = R_r + iR_i = 0$ . Now, what is the photon-tunneling equivalent of a black body? For  $q > \omega/c$  there are no constraints on the reflection coefficient  $R(q, \omega)$  other than that  $\text{Im} R(q, \omega)$  is positive. Therefore, assuming identical surfaces, we are free to maximize the transmission coefficient corresponding to the photon tunneling

$$T = \frac{R_i^2 e^{-2kd}}{|1 - e^{-2kd} R^2|^2} \quad (5)$$

This function is a maximum when [3]

$$R_r^2 + R_i^2 = e^{2kd} \quad (6)$$

so that  $T = 1/4$ . Substituting these results in (3) gives the evanescent contribution

$$(S_z)_{max}^{evan} = \frac{k_B^2 T^2 q_c^2}{24\hbar} \quad (7)$$

where  $q_c$  is a cut-off in  $q$ , determined by the properties of the material. It is clear that the largest possible  $q_c \sim 1/a$ , where  $a$  is an interatomic distance. Thus, from Eq. (7) we get upper boundary for the radiative heat transfer at room temperature:  $(S_z)_{max} \sim 10^{12} \text{Wm}^{-2}$ .

We rewrite the denominator of the integrand in the term in Eq. (3), which corresponding to the evanescent waves, in the form

$$|1 - e^{-2kd}R|^2 = [(1 - e^{-kd}R_r)^2 + e^{-2kd}R_i^2] \times [(1 + e^{-kd}R_r)^2 + e^{-2kd}R_i^2] \quad (8)$$

The conditions for resonant photon tunneling are determined by equation

$$R_r(\omega_{\pm}(q)) = \pm e^{kd} \quad (9)$$

This condition can be fulfilled even when  $\exp(-2kd) \ll 1$  because for evanescent electromagnetic waves there is no restriction on the magnitude of real part or the modulus of  $R$ . This open up the possibility of resonant denominators for  $R_r^2 \gg 1$ . Close to resonance we can write

$$(1 \pm e^{-kd}R_r)^2 \pm e^{-2kd}R_i^2 \approx e^{-2kd}R_r'^2(\omega_{\pm})[(\omega - \omega_{\pm})^2 + (R_i(\omega_{\pm})/R_r'(\omega_{\pm}))^2], \quad (10)$$

where

$$R_r'(\omega_{\pm}) = \left. \frac{dR_r(\omega)}{d\omega} \right|_{\omega=\omega_{\pm}},$$

which leads to the following contribution to the heat flux:

$$S_{\pm} \approx \frac{1}{4\pi} \int_0^{q_c} dk k (\Pi_1(\omega_{\pm}) - \Pi_2(\omega_{\pm})) \frac{R_i(\omega_{\pm})}{R_r'(\omega_{\pm})}. \quad (11)$$

The parameter  $q_c$  in this expression defines the region  $0 < q < q_c$  where the two- poles approximation is valid. To proceed further, let us make the following simplifications. Close to a pole we can use the approximation

$$R = \frac{a}{\omega - \omega_0 - i\eta}, \quad (12)$$

where  $a$  is a constant. Then from the resonant condition (9) we get

$$\omega_{\pm} = \omega_0 \pm ae^{-kd}.$$

For the two poles approximation to be valid the difference  $\Delta\omega = |\omega_+ - \omega_-|$  must be greater than the width  $\eta$  of the resonance. From this condition we

get  $q_c \leq \ln(2a/\eta)/d$ . For short distances the parameter  $q_c$  defines the value of  $q$  where the solution of Eq. (9) ceases to exist.

For  $\omega_0 > a$  and  $q_c d > 1$ , from Eq. (11) we get

$$J_{\pm} = \frac{\eta q_c^2}{8\pi} [\Pi_1(\omega_0) - \Pi_2(\omega_0)]. \quad (13)$$

Interesting, the explicit  $d$  dependence has dropped out of Eq. (13). However,  $J$  may still be  $d$ -dependent, through the  $d$ -dependence of  $q_c$ . For the small distances one can expect that  $q_c$  is determined by the dielectric properties of the material, and thus does not depend on  $d$ . In this case the heat transfer will be also distance independent.

### 3 Numerical results

Resonant photon tunneling enhancement of the heat transfer is possible for two semiconductor surfaces which can support low-frequency surface plasmon modes. The reflection factor  $R_p$  for clean semiconductor surface is given by Fresnel's formula

$$R_p = \frac{k - s/\epsilon}{k + s/\epsilon}, \quad (14)$$

where

$$s = \sqrt{k^2 - \left(\frac{\omega}{c}\right)^2 (\epsilon - 1)}, \quad (15)$$

where  $\epsilon$  is the bulk dielectric function. As an example, we consider two clean surfaces of silicon carbide (SiC). The optical properties of this material can be described using an oscillator model [16]

$$\epsilon(\omega) = \epsilon_{\infty} \left( 1 + \frac{\omega_L^2 - \omega_T^2}{\omega_T^2 - \omega^2 - i\Gamma\omega} \right) \quad (16)$$

with  $\epsilon_{\infty} = 6.7$ ,  $\omega_L = 1.8 \cdot 10^{14} s^{-1}$ ,  $\omega_T = 1.49 \cdot 10^{14} s^{-1}$ , and  $\Gamma = 8.9 \cdot 10^{11} s^{-1}$ . The frequency of surface plasmons is determined by condition  $\epsilon_r(\omega_p) = -1$  and from (16) we get  $\omega_p = 1.78 \cdot 10^{14} s^{-1}$ . In Fig.1 we plot the heat flux  $S(d)$ : note that the heat flux between the two semiconductor surfaces is several order of magnitude larger than between two clean good conductor surfaces (see Fig.3).

Another enhancement mechanism is connected with resonant photon tunneling between adsorbate vibrational modes localized on different surfaces. As an example, let us consider ions with charge  $e^*$  adsorbed on metal surfaces. The reflection factor  $R_p$ , which takes into account the contribution from an adsorbate layer, is given by [17]:

$$R_p = \frac{q - s/\epsilon + 4\pi n_a q [s\alpha_{\parallel}/\epsilon + q\alpha_{\perp}]}{q + s/\epsilon + 4\pi n_a q [s\alpha_{\parallel}/\epsilon - q\alpha_{\perp}]}, \quad (17)$$

and where  $\alpha_{\parallel}$  and  $\alpha_{\perp}$  are the polarizabilities of adsorbates in a direction parallel and normal to the surface, respectively.  $\epsilon$  is the bulk dielectric function and  $n_a$  is the concentration of adsorbates. For clean surfaces  $n_a = 0$ , and in this case formula (17) reduces to the Fresnel formula. The polarizability for ion vibration normal to the surface is given by

$$\alpha_{\perp} = \frac{e^{*2}}{M(\omega_{\perp}^2 - \omega^2 - i\omega\eta_{\perp})}, \quad (18)$$

where  $\omega_{\perp}$  is the frequency of the normal adsorbate vibration, and  $\eta_{\perp}$  is the damping constant. In Eq. (17) the contribution from parallel vibrations is reduced by the small factor  $1/\epsilon$ . However, the contribution of parallel vibrations to the heat transfer can nevertheless be important due to the indirect interaction of the parallel adsorbate vibration with the electric field, via the metal conduction electron [18]. Thus, the small parallel component of the electric field will induce a strong electric current in the metal. The drag force between the electron flow and the adsorbates can induce adsorbate vibrations parallel to the surface. This gives the polarizability:

$$\alpha_{\parallel} = \frac{\epsilon - 1}{n} \frac{e^*}{e} \frac{\omega\eta_{\parallel}}{(\omega_{\parallel}^2 - \omega^2 - i\omega\eta_{\parallel})} \quad (19)$$

where  $n$  is the conduction electron concentration. As an illustration, in Fig.2 we show the heat flux for the two Cu(001) surfaces covered by a low concentration of potassium atoms ( $n_a = 10^{18}m^{-2}$ ). In the  $q$ - integral in Eq.(3) we used the cut off  $q_c \sim \pi/a$  (where  $a \approx 1nm$  is the inter-adsorbate distance) because our microscopic approach is applicable only when the wave length of the electromagnetic field is larger than double average distance between the adsorbates. In comparison, the heat flux between two clean surface at separation  $d = 1nm$  is two order of magnitude smaller.



Fig.3 shows the thermal flux between two clean metal surfaces as a function of electron density  $n$ . In the calculations we have assumed that one body is at zero temperature and the other at  $T = 273$  K, and the Drude relaxation time  $\tau = 4 \cdot 10^{-14} \text{s}^{-1}$ . When the electron density decreases there is transition from a degenerate electron gas ( $k_B T \ll \varepsilon_F$ , where  $\varepsilon_F$  is the Fermi energy) to a non-degenerate electron gas ( $k_B T \gg \varepsilon_F$ ) at the density  $n_F \sim (K_B T m)^{3/2} / \pi^2 \hbar^3$ , where  $m$  is the electron mass. At  $T = 273$  K the transition density  $n_F \sim 10^{25} \text{m}^{-3}$ . The full line was obtained by interpolation between the two dashed curves, calculated in the non-local dielectric function formalism for the non-degenerate electron gas (valid for  $n < n_F \approx 10^{25} \text{m}^{-3}$ ), and for the degenerate electron gas (for  $n > n_F$ ) [13]. The thermal flux reaches the maximum  $S_{\text{max}} \approx 5 \times 10^8 \text{W} \cdot \text{m}^{-2}$  at  $n_{\text{max}} \approx 10^{25} \text{m}^{-3}$ , which corresponds to the DC conductivity  $\sigma \approx 3 \cdot 10^3 (\Omega \cdot \text{m})^{-1}$ . Within the local optic approximation the radiative heat transfer is maximal at  $n_{L \text{max}} \approx 10^{24} \text{m}^{-3}$  where  $S_{L \text{max}} \approx 10^9 \text{W} \cdot \text{m}^{-2}$ . The thermal flux due to traveling electromagnetic waves is determined by formula (1) which gives  $S_{BB} = 308 \text{W} \cdot \text{m}^{-2}$  for  $T = 273 \text{K}$ .

## 4 Summary

We have studied the radiative heat transfer in dependence on the dielectric properties of the media. We have found that at sufficiently short distances between bodies the thermal flux can be significantly enhanced in comparison with the black body radiation when the material involved support low-frequency adsorbate vibrational modes or surface plasmon modes, or the conductivity of the metals is chosen to optimize the heat transfer. This fact can be used in scanning probe microscopy for local heating and modification of surfaces.

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### FIGURE CAPTIONS

Fig. 1. The heat flux between two semiconductor surfaces as a function of separation  $d$ . One body is at zero temperature and the other at  $T = 273$  K. For parameters corresponding to a surface of silicon carbide (SiC) (see text

for the explanation). (The log-function is with basis 10)

Fig. 2. The heat flux between two surfaces covered by adsorbates, as a function of the separation  $d$ . One body is at zero temperature and the other at  $T = 273$  K. For parameters corresponding to K/Cu(001) [19] ( $\omega_{\perp} = 1.9 \cdot 10^{13} s^{-1}$ ,  $\omega_{\parallel} = 4.5 \cdot 10^{12} s^{-1}$ ,  $\eta_{\parallel} = 2.8 \cdot 10^{10} s^{-1}$ ,  $\eta_{\perp} = 1.6 \cdot 10^{12} s^{-1}$ ,  $e^* = 0.88e$ ) (The log-function is with basis 10)

Fig. 3. The heat flux between two metal surfaces as a function of the free electron concentration  $n$ . One body is at zero temperature and the other at  $T = 273$  K. The full line was obtained by interpolation between curves (dashed lines) calculated in the non-local dielectric formalism for a non-degenerate electron gas for  $n < n_F \sim 10^{25} m^{-3}$ , and for a degenerate electron gas for  $n > n_F$ . Also shown is results (dashed lines) obtained within the local optic approximation. The calculation were performed with the damping constant  $\tau^{-1} = 2.5 \cdot 10^{13} s^{-1}$ , separation  $d = 10 \text{ \AA}$  and  $n_0 = 8.6 \cdot 10^{28} m^{-3}$ . (The log-function is with basis 10)

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